Self-Polarization and Cooling of Spins in Quantum Dots

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Spontaneous nuclear polarization is predicted in double quantum dots in the spin-blocked electron transport regime. The polarization results from an instability of the zero-polarization state when singlet and triplet electron energy levels are brought into resonance by the effective hyperfine field of the nuclei on the electrons. The nuclear spins, once polarized, serve as a cold bath for cooling electrons below the lattice (phonon) temperature. We estimate the relevant time scales and discuss the conditions necessary to observe these phenomena.

Recent advances in semiconductor quantum dot technology have given experimentalists the ability to control the behavior of individual electrons and investigate their coupling to nuclear spins. Interesting phenomena such as switching, hysteresis, and long period oscillatory behavior of electric current were observed by Ono and Tarucha[1] in the so-called spin-blockade regime in GaAs vertical double quantum dots. Koppens et al. have also observed bistability and switching of electron current in lateral quantum dot system under analogous conditions [2]. In both cases, strong evidence was presented linking the observed phenomena to collective behavior of the nuclear spins in the lattice.

Collective behavior of nuclear spins in solids at Kelvin temperatures[1, 2], suggestive of spontaneous ordering, is a remarkable phenomenon since the natural dipole-dipole interaction between nuclear spins is so weak that microkelvin temperatures are required to achieve ordering under equilibrium conditions. The self-polarization of nuclei observed in Refs.[1, 2] was made possible by the non-equilibrium conditions of these experiments.

Much of recent theoretical work on spins in quantum dots has focused on dephasing [3, 4] and relaxation [5] of electron spins due to their interaction with the lattice nuclei. Additionally, Jouravlev and Nazarov[6] proposed a theory of spin-blockaded electron transport governed by the effective field of nuclear spins. In contrast to the idea of random fluctuations of the disordered nuclear spin system, in this work we are interested in the situation where spin-blockade leads to spontaneous ordering of a macroscopic fraction of the nuclear spin system.

Although qualitative scenarios explaining the above-mentioned instabilities have been offered[1, 2], a complete picture remains elusive. In a different context, however, Dyakonov and Perel studied instabilities involving nuclear spins in the optical pumping regime in bulk semiconductors[7]. Between this theoretical work and the unexplained observations of current-driven instabilities, there is a gap to be filled in.

In this paper we discuss a model of electron transport through a double quantum dot system which exhibits an instability toward self-polarization of the nuclei. In equilibrium at temperatures above a few millikelvin, nuclear spins are randomly oriented, having a root-mean-squared polarization of size $\mathcal{O}(\sqrt{N})$, where N is the total number of spins. To achieve an extensive polarization that scales with N, the system must be driven out of equilibrium.

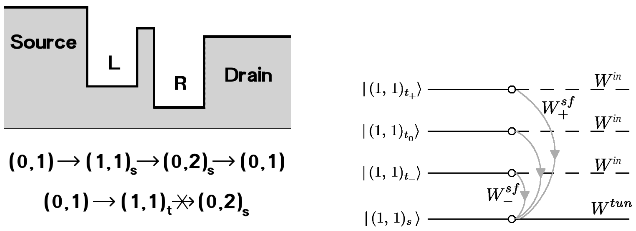
Polarization of nuclear spins occurs as an unpolarized electron current is passed through the double dot device. The hyperfine interaction between electrons and nuclei couples the dynamics of electron and nuclear spins. In a certain window of level detuning, feedback of the effective field of nuclear spins on electron levels near resonance becomes strong enough to pull the levels further into resonance by polarizing the nuclear spins. Transport drives the instability, taking the nuclear spins out of equilibrium and allowing an extensive polarization to develop. We discuss this effect and show that it can give rise to collective behavior of the otherwise independent nuclei.

Once one has successfully polarized the nuclear spins within a quantum dot, an interesting thermodynamic situation is obtained. Consider the equilibrium of a single electron spin isolated in such a quantum dot in the presence of an applied Zeeman field. While phonons are in equilibrium at the lattice temperature, the (non-equilibrium) nuclear subsystem has an effective temperature that is much lower. Assuming a static, non-equilibrium nuclear spin background, to what effective temperature does the electron spin relax?

The non-equilibrium nuclear spin distribution can affect the electron spin dramatically if electron spin relaxation is predominantly due to hyperfine spin flips. We compare the contributions to relaxation due to spin-orbital and hyperfine couplings, and conclude that the triplet states of a double dot system with two electrons can be cooled to an effective temperature $T_{\rm eff}$ below that of the phonon bath [see Eq. (10)].

In the paragraphs that follow, we outline the model put forth by Ono and Tarucha in [1] to explain the observed behavior of their vertical double quantum dot devices in the spin blockade regime. This model, accepted here for concreteness and to set the stage for our discussion, illustrates more general ideas that apply to a variety of systems with similar electron energy spectra.

The double dot is weakly coupled in series to two unpolarized leads, with transport occuring as a series of discrete hopping events as described in Figure 1(a). Current is suppressed when one of the three $(1,1)_t$ states are oc-



(a) Spin Blockade (b) Relevant Transitions

FIG. 1: (a) Schematic of the double quantum dot system. Initially the system is in state $(0,1)$ with one electron on the right dot. Current flows as depicted by the arrows: an electron tunnels from the source into the left dot to form the state $(1,1)_{s/t}$ with singlet or triplet spin, respectively. The electron then must hop to the right dot to form $(0,2)_s$ before tunneling out and returning the system to the $(0,1)$ state. If $(1,1)_t$ is formed when the left dot is filled, the Pauli Principle prohibits the second electron from tunneling into the right dot. (b) The relevant transitions included in our calculation. Bare spin flip rates $W^{\rm sf}_{\pm}$ are calculated by Eq.(1). Electrons can also escape from the dot without nuclear spin flip with rate $W^{\rm in}$, giving the net spin flip rate of the form (2).

cupied in the first step of the cycle. Once in the triplet state, residual current is due to slow indirect tunneling through virtual excited states, exchange with the leads, and spin-flips due to spin-orbit coupling and/or hyperfine flip-flop scattering. However, because spin-orbit effects are suppressed due to confinement in structures of this type [8, 9, 13, 14], we consider only the effects of indirect tunneling and hyperfine scattering.

For simplicity, we assume that tunneling rates from the weakly coupled source lead into the four $(1,1)_{s/t}$ states are all equal. Coupling of the $(0,2)_s$ state to the drain lead is stronger and gives this level a significant decay width. The finite width of this level plays a key role in the feedback mechanism that drives the polarization instability by favoring transitions from one of the split-triplet levels as it approaches resonance with $(0,2)_s$.

In general, the orbital eigenstates with singlet spin configuration are a superposition of the states $(1,1)_s$ and $(0,2)_s$. Here we assume, without loss of generality, that one eigenstate retains predominantly the $(1,1)$ charge distribution, while the other retains predominantly the $(0,2)$ charge distribution. It is these "$(1,1)_s$-like" and "$(0,2)_s$-like" states to which we refer below.

The transitions relevant to our model are depicted in Figure 1(b). We assume that nuclear dynamics are incoherent, and describe them by the populations $N_{\pm}$ of the up and down nuclear spin states [18]. We also neglect spatial variations in the nuclear spin population and transitions that do not change the net spin, which are inessential for our analysis. The energy-dependent hyperfine spin flip transition rates $W^{\rm sf}_{\pm}$ are calculated using Fermi's Golden Rule:

$$W^{\rm sf}_{\pm} = \frac{2\pi}{\hbar} |\langle\, (0,2)_s\, |\hat{H}_{\rm hf}|\, (1,1)_{t_\pm}\,\rangle|^2\, N_{\mp}\, f(\varepsilon_{t_\pm} - \varepsilon_s) \quad (1)$$

where $f(\varepsilon)$ is the density of states for the singlet final state, and $\varepsilon_{t_\pm}$, ε_s are the energies of the triplet and singlet, respectively. We assume a Lorentzian lineshape

$$f(\varepsilon) \propto \frac{\gamma}{(\varepsilon - \varepsilon_s)^2 + \gamma^{\ 2}}.$$

to allow explicit calculation.

Our use of Fermi's Golden Rule (1) is valid when nuclear spins dephase on the time scale of a few electron tunneling events. This situation is to be contrasted with the picture involving coherent hyperfine coupling discussed by Taylor et al. [16] which ignores nuclear spin dephasing and obtains typical polarization $\mathcal{O}(\sqrt{N})$.

When electrons are injected from an unpolarized source and every electron must exchange its spin with a nucleus to escape, no spin can be pumped into the nuclear spin system irrespective of the ratio of the rates for flipping nuclei up or down, $W^{\rm sf}_{+}/W^{\rm sf}_{-}$. If, on the other hand, electrons have an alternative way to escape, it need not be the case that the same number of nuclei must flip their spins in each direction.

For simplicity, we assume a single energy-independent indirect tunneling rate $W^{\rm in}$ for all three triplet states that relieves spin-blockade without interaction with the nuclei. Due to the competition between these processes, the net nuclear spin flip rates are given by

$$W^{\rm sf\prime}_{\pm} = \frac{I_0}{4} \frac{W^{\rm sf}_{\pm}}{W^{\rm sf}_{\pm} + W^{\rm in}}, \quad (2)$$

where I_0 is the total current through the system[19]. If the energy dependent rates $W^{\rm sf}_{\pm}$ are not equal, it is possible for electron transport to be dominated by spin-flip processes for electrons of one spin type, and by indirect tunneling processes for electrons of the other type.

The nuclear system is in a steady state when the rates $W^{\rm sf\prime}_{+}$ and $W^{\rm sf\prime}_{-}$ are equal. Assuming no dependence of the matrix elements containing the orbital wavefunctions on the electron spin-projection, we have (cf. Ref.[7]):

$$f(\varepsilon_+)\, N_- = f(\varepsilon_-)\, N_+, \quad \varepsilon_\pm = \varepsilon_{t_\pm} - \varepsilon_s. \quad (3)$$

When $f(\varepsilon_+) \neq f(\varepsilon_-)$, there can be a nonzero nuclear spin polarization in the steady state even when electron Zeeman energy is negligible. Let $x \equiv (N_+ - N_-)/N$ be the fractional nuclear polarization. Due to the hyperfine coupling-induced Overhauser shift, the triplet levels are split: $\varepsilon_\pm = \varepsilon_0 \pm \alpha N x$, where ε_0 is the detuning from the triplet-singlet resonance. For GaAs, where $g_e \approx -0.44$, $\alpha < 0$, this leads to the situation depicted in Fig.2.

The condition (3) yields a third order equation for the equilibrium polarization x^* with solutions

$$x^* = 0, \quad x^*_\pm = \frac{\sqrt{-2\varepsilon_0 \alpha N - ({\varepsilon_0}^2 + \gamma^2)}}{\alpha N}. \quad (4)$$

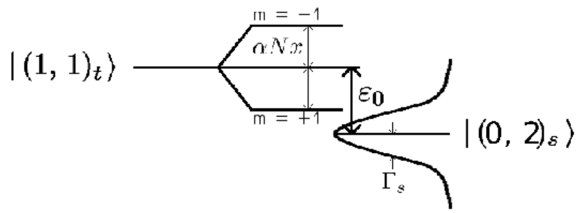


FIG. 2: Electron energy levels involved in the spin blockade. The $m = 0$ triplet level is detuned from the singlet by energy ε_0. When the nuclear spin system has fractional polarization x, the effective (Overhauser) field causes a splitting of $\alpha N x$ between the triplet levels. Resonance of one of the triplet levels with the broadened $(0,2)_s$ state favors spin flips for this state.

Which of these solutions corresponds to a stable equilibrium? The solution $x^* = 0$ always exists, for at $B_{\text{ext}} = 0$ the triplet levels are degenerate when $x = 0$. However, the solutions with finite nuclear polarization exist only when the discriminant is positive, i.e.

$$(\varepsilon_0 + \alpha N)^2 + \gamma^2 < (\alpha N)^2 \tag{5}$$

For a detuning of order γ, relation (5) requires that the electron hyperfine shift at full nuclear polarization is larger than the singlet level width. When this condition is met, the zero-polarization solution $x^* = 0$ is unstable.

Beginning at large detuning where (5) is not satisfied, the system stably remains at the equilibrium $x^* = 0$. This state loses stability once ε_0 is reduced past the threshold, and the system tends to one of the solutions $x^*_\pm$. At optimal detuning when $\varepsilon_0 = -\alpha N$, the spontaneous polarization reaches

$$N_+ - N_-|_{\max} = \pm\sqrt{N^2 - \gamma^2/\alpha^2}. \tag{6}$$

This should be contrasted with an $\mathcal{O}(\sqrt{N})$ polarization due to the initially randomized nuclear spins. In typical GaAs quantum dots, fully polarized nuclei can create an effective field of strength of up to a few Tesla, which translates into a triplet splitting of $\varepsilon_{t_+} - \varepsilon_{t_-} \approx 0.1$meV.

For a total current $I_0 \approx 1$pA, there will be one spin-flip for every few tunneling events leading to a net spin-flip rate of $\mathcal{O}(I_0/e) \approx 10^6 - 10^7 s^{-1}$. In a sample with $N = 10^5$ nuclei, this means that a particular nucleus is pumped with a rate $\mathcal{O}(I_0/eN) \approx 10 - 10^2 s^{-1}$. Nuclear relaxation is heavily suppressed at low temperatures, and can be assumed to take place on a much longer timescale than this. However, the nuclear dipole-dipole flip rate $W_{DD} \approx 10^3 s^{-1}$ (corresponding to a dipole field of $10^{-4} T$) is non-negligible on the predicted time scale of pumping and must not be ignored.

The dipole-dipole interaction does not conserve total spin due to the presence of terms such as $\hat{S}^z_i \hat{S}^+_j$, $\hat{S}^+_i \hat{S}^+_j$, etc. (e.g., see [10]). In a weak applied field, the Zeeman splitting of nuclear spin states with different total z-projection quenches the action of such terms due to energy nonconservation. The strength of the applied field required for that need only give rise to Zeeman splitting greater than the dipole-dipole level width. Typically, a field $B_{\text{ext}} \approx 0.5$mT is sufficent for this purpose.

The terms remaining after application of the small quenching field allow nuclear spin to diffuse throughout space (see Ref.[10] for discussion of spin diffusion). Using experimentally determined diffusion parameters [11, 12], we estimate a characteristic time of 10 seconds for diffusion out of the dot. This rate is several orders of magnitude smaller than the rate of pumping and does not inhibit the pumping of nuclear spins.

We now discuss the cooling of electron spins by the polarized nuclear spin bath. Due to the small nuclear Zeeman energy compared to that of an electron, hyperfine-induced electron spin relaxation must involve additional degrees of freedom such as phonons to carry away the excess energy. Electron spin relaxation in a random nuclear field in a single quantum dot has been studied by Erlingsson, Nazarov, and Fal'ko [5]. Spin-orbit relaxation rates were studied by Khaetskii and Nazarov [8] and by Golovach, Khaetskii, and Loss [9]. These estimates along with recent experimental results [13] indicate that on time scales of interest the rate of spin relaxation for a single electron is dominated by the spin orbit mechanism with phonon emission.

In contrast, Johnson et al. found that relaxation of a two-electron triplet state in a double quantum dot was dominated by hyperfine interactions at magnetic fields less than approximately 100 mT [15]. The reason for this reversal of roles is that hyperfine transitions are enhanced by the near resonance of singlet and triplet levels in the double dot system, whereas the spin orbit coupling is suppressed by a small tunneling matrix element. Thus in this low-field regime, the presence of a polarized nuclear spin bath may cause the electron spin degrees of freedom to adopt an equilibrium distribution different from the thermal distribution at the lattice temperature [20].

We estimate the cooling effect for the system considered above, with couplings to the leads tuned to zero and a gating bias favoring the unbroadened $(0,2)_s$ state. As before, we treat transitions as incoherent using Fermi's Golden Rule, this time via second order perturbation theory in the hyperfine and electron-phonon couplings. At this level the transitions with $\Delta m = \pm 2$ are not allowed. Rates for all other "forward" and "reverse" processes along with the normalization condition $\sum P_k = 1$ yield a system of five linear equations in the five unknown equilbrium populations P_k, $k = 0, ..., 4$ stands for $(0,2)_s$, $(1,1)_{t_+}$, $(1,1)_{t_0}$,$(1,1)_{t_-}$, or $(1,1)_s$, respectively.

The rate equations governing the relaxation dynamics of this system are given by

$$\dot{P}_k = \sum_j (\Gamma_{kj} P_j - \Gamma_{jk} P_k) \tag{7}$$

where Γ_{kj} is the transition rate from state j to state k. For example, the relaxation rate Γ_{01} from $(1,1)_{t_+}$ to

$(0,2)_s$ via virtual transition through $(1,1)_s$ is

$$\Gamma_{01} = \Gamma_{\rm ph}[\Delta E_{01}]\ (\mathcal{M}/\Delta E_{41})^2\ N_-, \tag{8}$$

derived as in Ref.[5] for the case of a single dot. Here $\Delta E_{ij} = E_i - E_j$ and $\Gamma_{\rm ph}[\Delta E]$ is the rate of phonon absorption/emission for positive/negative energy ΔE.

The phonon rates $\Gamma_{\rm ph}[\Delta E]$ in equation (8) obey the detailed balance relation $\Gamma_{\rm ph}[\Delta E_{jk}]/\Gamma_{\rm ph}[\Delta E_{kj}] = e^{-\beta_{\rm ph}\Delta E_{jk}}$, with $T_{\rm ph} = \beta_{\rm ph}^{-1}$ the phonon bath temperature. The ratios of forward and reverse rates for transitions that include a change of m, however, include an additional factor of $N_-/N_+ \equiv e^{-\beta_n E_n}$ or its reciprocal. This relation defines the effective temperature $T_n = \beta_n^{-1}$ of the nuclear subsystem, with $E_n = \mu_n B$ the Zeeman energy of a single nucleus. Here μ_n is the nuclear magnetic moment and B is the applied magnetic field strength.

In the steady state with $\dot{P}_k = 0$ for all k, the populations obtained from equation (7) obey

$$\begin{array}{ll} P_2/P_0 = e^{-\beta_{\rm ph}\Delta E_{20}}, & P_2/P_1 = e^{-\beta_{\rm ph}\Delta E_{21}}\, e^{-\beta_n E_n}, \\ P_4/P_0 = e^{-\beta_{\rm ph}\Delta E_{40}}, & P_3/P_2 = e^{-\beta_{\rm ph}\Delta E_{32}}\, e^{-\beta_n E_n}. \end{array} \tag{9}$$

These relations describe a non-thermal distribution such that the energy dependence (9) cannot be accounted for by a single effective temperature $T_{\rm eff} = \beta_{\rm eff}^{-1}$.

Non-Boltzmann aspects aside, this process can still be considered as a form of cooling as a finite (positive) nuclear spin temperature leads to a transfer of population from high energy to low energy states. The energy levels within the triplet subspace are evenly spaced with $\Delta E_{21} = \Delta E_{32} = \mu_{el} B$, where μ_{el} is the electron magnetic moment. This leads to a quasi-thermal distribution within this subspace: $e^{-\beta_{\rm eff}\Delta E_{21}} \equiv P_2/P_1 = P_3/P_2 = e^{-\beta_{\rm ph}\Delta E_{21}}\, e^{-\beta_n E_n}$, with effective temperature

$$T_{\rm eff} = \frac{T_n T_{\rm ph}}{T_n + (\mu_n/\mu_{el}) T_{\rm ph}} \tag{10}$$

with $\mu_n/\mu_{el} \approx 10^{-3}$. For a lattice temperature of 100 mK and total field of 100 mT, $T_{\rm eff}$ will differ significantly from T for nuclear polarizations greater than about 50%.

To summarize, the key ingredient that makes it possible to generate a finite nuclear polarization using current from *unpolarized* leads is the competition between energy dependent spin-flip and energy independent spin-conserving tunneling channels. When spin-flips are favored in one spin state but spin-conserving tunneling favored for the other, polarization will be pumped into the nuclear spin system. Once a non-equilibrium nuclear spin state is established, it can be used as a cold bath to cool electron spins below the lattice temperature. While relatively fast spin-orbit processes in GaAs prohibit cooling of a single electron spin, two-electron states of a double quantum dot are amenable to such cooling.

Electron transition dependence on Overhauser shift as a cause of bi-stability in spin-blockaded transport was discussed by Inarrea, Platero and MacDonald in Ref.[17], which appeared after this work was completed.

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[18] Although our primary example is GaAs which is comprised of spin-3/2 nuclei, for clarity of presentation we proceed with the calculation assuming spin-1/2 nuclei. This allows us to introduce the populations of nuclei in the up and down spin states, N_+ and N_-.
[19] In principle the current can be found self-consistently from the rates of the individual processes. The energy and nuclear-spin state dependence of the current leads to a quantitative change in the rate of spin pumping, but does not affect our general conclusions.
[20] Although the Overhauser field of a nearly polarized nuclear system is of order 1 T, an opposing external field can be applied to make the total effective field seen by the electrons fall into the low-field regime.